Heat capacity of a thin membrane at very low temperature

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We calculate the dependence of heat capacity of a free standing thin membrane on its thickness and temperature. A remarkable fact is that for a given temperature there exists a minimum in the dependence of the heat capacity on the thickness. The ratio of the heat capacity to its minimal value for a given temperature is a universal function of the ratio of the thickness to its value corresponding to the minimum. The minimal value of the heat capacitance for given temperature is proportional to the temperature squared. Our analysis can be used, in particular, for optimizing support membranes for microbolometers.

a. Introduction: Thin free standing membranes are extensively used for sensing and detecting, in particular, for mounting of microbolometers.[1] Thermal and heat transport properties of such membranes are very important for sensitivity of such bolometers and their time response. At low temperatures the wavelength of thermal phonons responsible for the heat capacity and conductance can exceed the membrane thickness, b. In this case the vibrational modes significantly differ from those in bulk materials.[2] In particular, the lowest vibrational mode has a *quadratic* rather than linear dispersion law.[3]

The contributions of these modes to the low-frequency density of vibrational states *increase* with decrease of the membrane thickness. As a result, the low-temperature heat capacity also *increases*. As the thickness increases, the heat capacity is determined by higher modes having an essentially linear dispersion law. Consequently, the heat capacity crosses over to that of a bulk material. As a result, the thickness dependence of the heat capacity of thin membranes is *nonmonotonous* having a sharp minimum at some optimal, temperature dependent thickness. This minimum is similar to the minimum in thickness dependence of the ballistic heat transfer (power radiation) predicted in Ref. 4. However, the concrete values of the optimal thickness for the heat capacity and the ballistic heat transfer are different. Consequently, a proper choice of the membrane thickness can be used for optimizing of the time response of microbolometers mounted on thin free standing membranes. The present paper is aimed at the theory of low temperature heat capacity of thin free standing membranes.

b. Vibrational spectrum: The vibrational modes of a thin membrane are superpositions of bulk longitudinal and transverse modes, their relative weights being determined by boundary conditions – both normal and tangential stresses should vanish. The eigenmodes are classified as symmetric (SM) and antisymmetric (AM). Both are superpositions of the longitudinal and transverse bulk modes with wave vectors $\mathbf{k}^l$ and $\mathbf{k}^t$, respec-

tively. The relations between $\mathbf{k}^l$ and $\mathbf{k}^t$ are[3]

$$\frac{\tan(bk_\perp^t/2)}{\tan(bk_\perp^l/2)} = -\frac{4k_\perp^l k_\perp^t k_\parallel^2}{[(k^t)_\perp^2 - k_\parallel^2]^2}, \tag{1}$$

$$\frac{\tan(bk_\perp^l/2)}{\tan(bk_\perp^t/2)} = -\frac{4k_\perp^l k_\perp^t k_\parallel^2}{[(k_\perp^t)^2 - k_\parallel^2]^2} \tag{2}$$

for SM and AM, respectively. Here $k_\perp$ and $k_\parallel$ denote perpendicular and parallel components of the wave vectors with respect to the membrane. Inserting the dispersion laws of the bulk modes, $k^l = \omega/c_l$ and $k^t = \omega/c_t$ where c_l and c_t are speeds of transversal and longitudinal sound, into Eqs. (1) and (2) one obtains transcendental equations for the dispersion laws, $\omega_{s,n}(k_\parallel)$, of different vibrational branches. Here the subscript σ stands for the branch type while n stands for its number.

In addition to AM and SM there exists a horizontal shear mode (HS), which is a transversal wave with both displacement and wave vector parallel to the plane of the membrane. The HS mode dispersion law is[3]

$$\omega_{\text{HS},n} = c_t\sqrt{(n\pi/b)^2 + k_\parallel^2} \tag{3}$$

where n is an integer number. SM, AM and HS modes are the only vibrations that can exist in a membrane. The modes possess a very important property: as follows from Eqs. (1), (2), and (3), the frequencies of all modes scale as[1]

$$\omega_{s,n} = 2c_t b^{-1} w_{s,n}(bk_\parallel). \tag{4}$$

Figure 1 shows the six lowest branches. Only the three lowest branches are gapless, consequently only they contribute to the heat capacity of a membrane at $k_B T \ll \hbar\omega_{\text{HS},1}(0)$. The lowest HS and SM are linear at $k \ll b^{-1}$ while the AM dispersion law can be approximated at small $k_\parallel$ as[2]

$$\omega_{\text{AM},0}(k_\parallel) = \frac{\hbar}{2m^*}k_\parallel^2, \quad m^* \equiv \frac{\hbar c_l\sqrt{3}}{2c_t b\sqrt{c_l^2 - c_t^2}}. \tag{5}$$
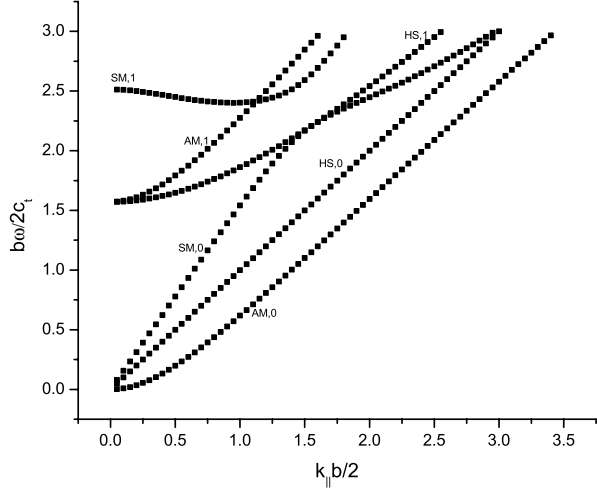
FIG. 1: The six lowest branches of vibrations in a membrane. The lowest AM branch is quadratic around zero.

The lowest AM branch is a flexural wave and the dispersion law of this branch can be obtained from the 2D analog of Bernoulli-Euler theory.[5] We will reproduce the derivation here since we will need it for analysis of the applicability range of our theory. Choosing the z-axis perpendicular to the membrane we have:[5]

$$\rho\frac{\partial^2 u}{\partial t^2} + \frac{D}{b}\Delta^2 u = 0\,, \quad D = \frac{b^3 E}{12(1-\sigma^2)}. \qquad (6)$$

where u is the displacement perpendicular to the membrane, ρ is the density of the membrane material, E is the Young modulus, σ is the Poisson ratio and Δ is the 2D Laplace operator. Searching for a solution in the form $u \sim e^{i(\mathbf{k}_\parallel \mathbf{r} - \omega t)}$ we obtain the dispersion law for flexural waves:

$$\omega_{fl} = k_\parallel^2 \left(D/\rho b\right)^{1/2}\,. \qquad (7)$$

Substituting the conventional expressions for the Young modulus and Poisson ratio through the sound velocities c_l and c_t and density ρ we arrive at the same dispersion law as given by Eq. (5). This equation is valid only in the long wave approximation, $\lambda \gg b$.

c. *Heat capacity of a free standing membrane:* We compute the heat capacity per unit area from the general equation

$$C = \frac{1}{A} \sum_{s,n,\mathbf{k}_\parallel} \frac{(\beta\hbar\omega_{s,n(\mathbf{k}_\parallel)})^2 e^{\beta\hbar\omega_{s,n}(\mathbf{k}_\parallel)}}{[e^{\beta\hbar\omega_s(\mathbf{k}_\parallel)}-1]^2}\,, \quad \beta \equiv \frac{1}{k_B T} \qquad (8)$$

where A is the area of the membrane.

The heat capacity per unit area C of a thick membrane ($b \gg \hbar\pi\beta c_t$) increases linearly with the thickness b as it should for a 3D body. But for a thin membrane ($b < \hbar\pi\beta c_t$) there is a minimum of heat capacity. The

position of this minimum depends not only on the properties of the membrane material but also on temperature. It is important to underline that we calculated the heat capacity of a membrane per unit area. To find the specific heat capacity c_v we should divide the heat capacity per unit area C by the thickness b.

For temperature $T \ll \hbar\pi c_t/k_B b$ the heat capacity of a membrane is dominated by the contribution of the lowest AM branch. So we can find a crude estimate for the position of the minimum, $b_{\min}$, by equating the heat capacity per unit area of a bulk sample to the contribution of the lowest AM branch to the heat capacity per unit area of a thin film.

We find the total heat capacity per unit area of a bulk sample by multiplying the thickness b of the membrane and its specific heat capacity c_v,[6]

$$\frac{c_v}{k_B} \approx \frac{2\pi^2}{5}\left(\frac{k_B T}{\hbar c}\right)^3\,, \quad \frac{1}{c^3} \equiv \frac{1}{3}\left(\frac{2}{c_t^3} + \frac{1}{c_l^3}\right)\,. \qquad (9)$$

The contribution of the lowest AM with the dispersion law (5) can be easily calculated from Eq. (8) as

$$\frac{C_{\mathrm{AM},0}}{k_B} = \frac{k_B T m^* \zeta(2)}{\pi\hbar^2}\,. \qquad (10)$$

From the equality $c_v b = C_{\mathrm{AM},0}$ we obtain

$$b_{\min}T = a\frac{c_l \hbar^2 c^3}{k_B^2 c_t\sqrt{c_l^2 - c_t^2}}\,, \quad a = \frac{5\sqrt{3}\zeta(2)}{4\pi^3} \approx 0.11\,. \qquad (11)$$

For the temperature 0.1 K the estimated position of the heat capacity minimum $b_{\min} \approx 400$ nm.

A more accurate procedure is based on the exact expression (8). Making use of the scaling relation (4) one can cast this equation in the form

$$C = \frac{k_B}{2\pi b^2}\mathcal{F}\left(\frac{\hbar c_t}{b k_B T}\right)\,, \qquad (12)$$

$$\mathcal{F}(z) \equiv \sum_{s,n}\int_0^\infty \xi\,d\xi\,\frac{[zw_{s,n}(\xi)]^2}{\sinh^2[zw_{s,n}(\xi)]}\,. \qquad (13)$$

Here $\xi \equiv bk_\parallel$ and we assume that the temperature is well below the Debye temperature so that we can integrate from 0 to infinity. One sees that the heat capacity per unit area is a nonmonotonous function of the film thickness, b. Its minimum can be determined by equating the derivative $\partial C/\partial b$ to zero. It leads to the relation

$$b_{\min} \cdot T = \frac{\hbar c_t}{k_B z^*} \qquad (14)$$

where z^* is determined from the equation $\mathcal{F}'(z^*)z^* = -2\mathcal{F}(z^*)$. The value of z^* depends on the ration c_t/c_l and for $c_t/c_l = 1.6$, $z^* \approx 1.9$.

Numerical calculations including the 30 lowest branches give the value $b_{\min} = 240$ for $T = 0.1K$, which
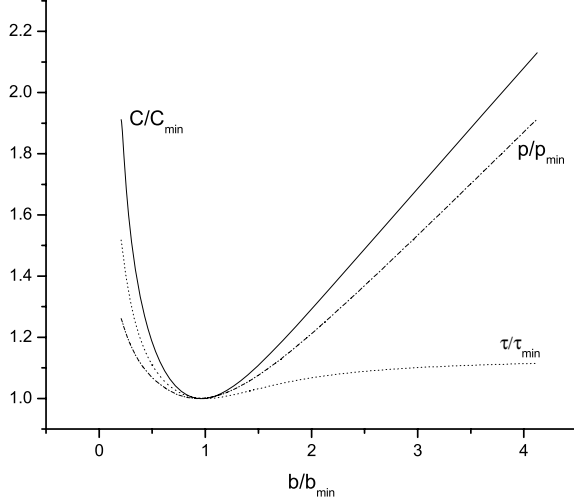
FIG. 2: The heat capacity per unit area C of a membrane, the response time τ and the derivative of the radiation power with temperature p as a function of its thickness b. $b_{\min}$ is the thickness for which the heat capacity reaches its minimal value. We used the values $c_t = 6000$ m/s, $c_l/c_t = 1.6$, which are approximately the same as in the experiments[1] on SiN_x membranes.

is not far from the crude estimate obtained using only the lowest mode. Substituting (14) into Eq. (12) we obtain

$$C_{\min} = \frac{(k_B T z^*)^2}{2\pi(\hbar c_t)^2} k_B \mathcal{F}(z^*) \,. \qquad (15)$$

The result can be summarized in the universal form

$$\frac{C}{C_{\min}} = \left(\frac{b_{\min}}{b}\right)^2 \frac{\mathcal{F}(z^* b_{\min}/b)}{\mathcal{F}(z^*)} \qquad (16)$$

where $b_{\min}(T)$ is given by Eq. (14). This is shown in Fig. 2.

d. *Applicability range of the theory:* There are several phenomena limiting the increase of the heat capacity with decrease of the film thickness. The obvious limit is that the elasticity theory is not applicable when the thickness is comparable with interatomic distance. Below we will discuss another limit posed by anharmonic effects. The amplitude of thermal vibrations increases with decreasing thickness. As a result, in a very thin membrane the harmonic approximation used in this paper becomes invalid.

To make an estimate, let us assume for simplicity that thickness of the membrane $b \ll b_{\min}$ and take into account only the AM,0 branch. Due to the thermal vibrations, the membrane will not be perfectly flat but have a shape described by some function $f(x,y)$. The resulting increase in the area of the membrane corresponds to some average tension, $\mathcal{T}$, of the membrane. Let us therefore first study the membrane in the presence of an external tension, and later insert the thermal value of this tension.
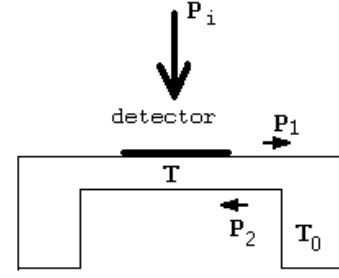

FIG. 3: Schematic of a free membrane with mounted detector.

In this case in the equation of motion (6) an additional term appears:

$$\rho\frac{\partial^2 u}{\partial t^2} = -\frac{Eb^2}{12(1-\sigma^2)}\frac{\partial^4 u}{\partial x^4} + \mathcal{T}\frac{\partial^2 u}{\partial x^2}\,, \qquad (17)$$

the dispersion law being

$$\omega = \sqrt{\frac{Eb^2}{12\rho(1-\sigma^2)}k^4 + \frac{\mathcal{T}}{\rho}k^2}\,. \qquad (18)$$

When the second term in (17) becomes greater than the first one the dispersion law changes to linear and the heat capacity of the membrane approaches some limiting value. Since thermal vibration produce an average strain they will change the dispersion law. So for a crude estimate we will evaluate the average strain produced by the thermal vibrations and compare its contribution with the first item $Eb^2k^4/12\rho(1-\sigma^2)$. We can express variation of the membrane area as $\Delta A \approx \frac{1}{2}\int_A |\nabla f|^2\,dxdy$. Substituting for f thermal modes of a square membrane and performing thermal average one obtains an estimate for the typical elongation:

$$\frac{\Delta L}{L} \approx \frac{k_B T}{\rho b^3(c_l^2 - c_t^2)}\frac{c_l^2}{c_t^2}\ln\frac{L}{L_T}\,, \quad L_T = \sqrt{\frac{2\pi^2\hbar^2}{m^* k_B T}}\,. \quad (19)$$

This relative elongation strongly depends on the membrane thickness. Substituting $\mathcal{T} \approx E(\Delta L/L)$ in Eq. (17) and comparing its contribution with the first item for thermal wave vectors we obtain an estimate of the critical thickness below which out theory is not applicable:

$$b_{cr} = \left(\frac{12\hbar c_l}{c_t\sqrt{3(c_l^2 - c_t^2)\rho}}\cdot\ln\frac{L}{L_T}\right)^{1/4}\,. \qquad (20)$$

Numerically, this estimate gives the thickness of several atomic layers, so it does no pose additional limitations comparing to general range of applicability of the elasticity theory.

e. *Sensitivity and response time of the bolometer:* Suppose we use a thin free standing membrane with a detector on it as a sensitive element of a bolometer. In

presence of incident flow of energy, P_i, see Fig. 3, we can obtain the temperature of a membrane as a function of time:

$$P_i(t) = P_1(T) - P_2(T_0) + C_{mem}(T)(dT/dt) \qquad (21)$$

where C_{mem} is the heat capacity of the membrane under the detector, $P_2(T_0)$ is the heat flow from surrounding media to the membrane under the detector, $P_1(T)$ is the heat flow from the membrane under the detector and T_0 is the temperature of surrounding media. We assume that phonons are ballistic,[4] i. e., the mean free path of the phonons is much longer than the size of the detector. Usually the relative temperature difference is less than 0.01, so we can expand the radiation power $P_1(T)$ around T_0 and use heat capacity as a constant. In this case we obtain a response time $\tau = C/(\partial P/\partial T)$. In equation (12) the part that depends only on bT can be separated

$$C = T^2 \tilde{C}(bT) \qquad (22)$$

The equation for power radiation[4] can be similarly transformed to

$$P = T^3 \tilde{P}(bT) . \qquad (23)$$

From this we can obtain response time is a function only of bT

$$\tau = \frac{\tilde{C}(bT)}{3\tilde{P}(bT) + bT\tilde{P}'(bT)} . \qquad (24)$$

From this we immediately obtain that the minimal response time, τ_{min}, as a function of membrane thickness does not depend on temperature. The dependence of the response time τ versus the membrane thickness is shown on the Fig. 2. The position of τ_{min} is close to the position of C_{min}. On Fig. 2 this difference is difficult to see because it is only 5%. If we measure pulses of energy shorter than τ the rate of temperature increase depends only on the heat capacity of the membrane. In the opposite case when pulses are longer than τ the final temperature difference depends only on $p = (\partial P/\partial T)$. The dependence of p on the ratio b/b_{min} is also presented on Fig. 2. From this picture we can see that the heat capacity C, the derivative of the radiation power with temperature p and response time τ reach their minimal values at different thicknesses. This difference is about 10%. If one want to construct a detector he wants to obtain either the shortest response time or the greatest temperature difference. Our calculations demonstrate that the best sensitivity and the shortest response time can be obtained by choosing the supporting membrane with thickness b_{min}.

f. Discussion and conclusions: We have studied the heat capacity of a thin membrane at low temperatures such that the typical wavelength of thermal phonons is of the order or smaller than the thickness of the membrane. Because of the quadratic dispersion law of the lowest vibrational branch, the heat capacity per area will increase with decreasing thickness below a certain thickness b_{min}. The thickness of minimal heat capacity is temperature dependent, $b_{min} \propto 1/T$. The shape of the curve $C(b)$ has the universal form (15). If we want to use the membrane for support of microbolometers, the reduction of the heat capacity is important for sensitivity of the detector for short pulses, and the thickness should be chosen equal to b_{min} at the operating temperature of the bolometer.

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